

Application No. 10/099,597

AMENDMENTS TO THE CLAIMS

A detailed listing of all claims that are, or were, in the present application, irrespective of whether the claim(s) remains under examination in the application are presented below. The claims are presented in ascending order and each includes one status identifier. Those claims not cancelled or withdrawn but amended by the current amendment utilize the following notations for amendment: 1. deleted matter is shown by strikethrough for six or more characters and double brackets for five or less characters; and 2. added matter is shown by underlining.

- 06 to extra X
1. (Original) A collection of amorphous particles comprising non-rare earth metal/metalloid host composition and a rare earth metal dopant/additive, the collection of particles having an average primary particle diameters less than about 500 nm.
 2. (Original) The collection of amorphous particles of claim 1 wherein the primary particles have an average diameter less than about 250 nm.
 3. (Original) The collection of amorphous particles of claim 1 wherein the primary particles have an average diameter less than about 100 nm.
 4. (Original) The collection of amorphous particles of claim 1 wherein the particles comprise from about 0.01 mole percent to about 10 mole percent rare earth metal dopant/additive composition content relative to the total host composition and dopant/additive composition content.
 5. (Original) The collection of amorphous particles of claim 1 wherein the particles comprise from about 0.025 mole percent to about 5 mole percent rare earth metal dopant/additive

Application No. 10/099,597

composition content relative to the total host composition and dopant/additive composition content.

6. (Original) The collection of amorphous particles of claim 1 wherein the particles comprise from about 0.1 mole percent to about 3 mole percent rare earth metal composition content relative to the total host composition and dopant/additive composition content.

7. (Original) The collection of amorphous particles of claim 1 wherein the metal/metalloid host composition comprises a host oxide selected from the group consisting of TiO_2 , SiO_2 , GeO_2 , Al_2O_3 , P_2O_5 , B_2O_3 , TeO_2 and combinations thereof, and wherein the particles comprise at least about 40 mole percent of the host oxide relative to the total host composition and dopant/additive composition content.

8. (Original) The collection of particle of claim 1 wherein the host oxide comprises SiO_2 .

9. (Original) The collection of amorphous particles of claim 7 further comprising a non-rare earth metal dopant/additive selected from the group consisting of Ga, Mg, Sr, Ti, Si, Ge, Al, P, B, Te, Bi, Sb, La, Y, Zr, Pb, Li, Na, K, Ba, Zn, W, Ca, and combinations thereof.

10. (Original) The collection of amorphous particles of claim 9 wherein the particles comprise from about 0.05 mole percent to about 5 mole percent non-rare earth metal dopant/additive composition content relative to the total host composition and dopant/additive composition content.

11. (Original) The collection of amorphous particles of claim 1 wherein the rare earth metal dopant/ additive is selected from the group consisting of Ho, Eu, Ce, Tb, Dy, Er, Yb, Nd, La, Y, Pr, Tm and combinations thereof.

Application No. 10/099,597

12. (Original) The collection of amorphous particles of claim 1 wherein effectively no primary particles have a diameter greater than about 5-times the average diameter of the primary particles.

13. (Original) The collection of amorphous particles of claim 1 wherein effectively no primary particles have a diameter greater than about 3 times the average diameter of the primary particles.

14. (Original) The collection of amorphous particles of claim 1 wherein the primary particles have a distribution of particle diameters wherein at least about 95 percent of the primary particles have a diameter greater than about 45 percent of the average diameter of the primary particles and less than about 200 percent of the average diameter.

15. (Original) The collection of amorphous particles of claim 1 wherein the particles comprise at least five different metal/metalloid elements.

16. (Original) A collection of amorphous particles comprising a metalloid oxide selected from the group consisting of B_2O_3 and TeO_2 , and a metal/metalloid dopant/additive, the collection of particles having an average diameter no more than about 1000 nm, wherein the particles comprise at least about 51 mole percent metalloid oxide content relative to the total metalloid oxide and dopant/additive composition content of the particles.

Application No. 10/099,597

17. (Original) The collection of amorphous particles of claim 16 wherein effectively no primary particles have a diameter greater than about 5 times the average diameter.
18. (Original) The collection of amorphous particles of claim 16 wherein the primary particles comprise a distribution of particle diameters wherein at least about 95 percent of the primary particles have a diameter greater than about 45 percent of the average diameter and less than about 200 percent of the average diameter.
19. (Original) A collection of particles comprising a metalloid oxide selected from the group consisting of B_2O_3 and TeO_2 , the collection of particles having an average diameter no more than about 250 nm.
20. (Original) The collection of particles of claim 19 wherein the particles further comprise a metal/metalloid dopant/additive.
21. (Original) The collection of particles of claim 20 wherein the particles comprise at least about 51 mole percent metalloid oxide content relative to the total metalloid oxide and dopant/additive composition content of the particles.
22. (Original) The collection of particles of claim 20 wherein the metal/metalloid dopant/additive comprises a rare earth metal.

Application No. 10/099,597

23. (Original) The collection of particles of claim 20 wherein the metal/metalloid dopant/additive comprises a non-rare earth metal.

24. (Original) The collection of particles of claim 19 having an average diameter no more than about 150 nm.

25. (Original) The collection of particles of claim 19 having an average diameter from about 3 nm to about 100 nm.

26. (Original) The collection of particles of claim 19 wherein the metalloid oxide comprises B_2O_3 .

27. (Original) The collection of particles of claim 19 wherein the metalloid oxide comprises TeO_2 .

28. (Original) The collection of particles of claim 19 wherein effectively no primary particles have a diameter greater than about 5 times the average diameter.

29. (Original) The collection of particles of claim 19 wherein the primary particles comprise a distribution of particle diameters wherein at least about 95 percent of the primary particles have a diameter greater than about 45 percent of the average diameter and less than about 200 percent of the average diameter.

Application No. 10/099,597

30. (Original) A collection of amorphous particles comprising GeO_2 and a metal/metalloid dopant/additive, the collection of particles having an average diameter no more than about 500 nm.

31. (Original) The collection of amorphous particles of claim 30 wherein the collection of particles comprises at least about 30 weight percent GeO_2 .

32. (Original) The collection of amorphous particles of claim 30 wherein the collection of particles comprises at least about 51 weight percent GeO_2 .

33. (Original) The collection of amorphous particles of claim 30 wherein effectively no primary particles have a diameter greater than about 5 times the average diameter.

34. (Original) The collection of amorphous particles of claim 30 wherein the primary particles comprise a distribution of particle diameters wherein at least about 95 percent of the primary particles have a diameter greater than about 45 percent of the average diameter and less than about 200 percent of the average diameter.

35-128 (Cancelled)

Application No. 10/099,597

Please add the following new claims:

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129. (New) The collection of amorphous particle of claim 16 wherein the dopant comprises a rare earth metal.
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130. (New) The collection of amorphous particles of claim 16 wherein the dopant comprises Ga, Mg, Sr, Ti, Si, Ge, Al, P, B, Te, Bi, Sb, La, Y, Zr, Pb, Li, Na, K, Ba, Zn, W, Ca, or combinations thereof.
- 37
131. (New) The collection of amorphous particles of claim 16 wherein the metalloid oxide is B_2O_3 .
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132. (New) The collection of amorphous particles of claim 16 wherein the metalloid oxide is TeO_2 .
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133. (New) The collection of amorphous particles of claim 16 wherein effectively no primary particles have a diameter greater than about 3 times the average diameter.
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134. (New) The collection of amorphous particles of claim 16 having an average diameter of less than about 100 nm.
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135. (New) The collection of amorphous particle of claim 30 wherein the dopant comprises a rare earth metal.

Application No. 10/099,597

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~~136.~~ (New) The collection of amorphous particles of claim 30 wherein the dopant comprises Ga, Mg, Sr, Ti, Si, Al, P, B, Te, Bi, Sb, La, Y, Zr, Pb, Li, Na, K, Ba, Zn, W, Ca, or combinations thereof.

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~~137.~~ (New) The collection of amorphous particles of claim 30 wherein effectively no primary particles have a diameter greater than about 3 times the average diameter.

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~~138.~~ (New) The collection of amorphous particles of claim 30 having an average diameter of less than about 100 nm.

hydrolysis deposition has become the leader for commercial implementation of planar waveguides. Flame hydrolysis and forms of chemical vapor deposition have also been successful in the production of glass fibers for use as fiber optic elements. Flame hydrolysis deposition involves the use of a hydrogen-oxygen flame to react gaseous precursors to form particles of the optical material as a coating on the surface of the substrate. Subsequent heat treatment of the coating can result in the formation of a uniform optical material, which generally is a glass material.

Substantially uniform coatings, e.g., glass coatings, can be incorporated into optical structures, such as planar optical structures. Planar optical structures can comprise integrated optical devices. Material processing remains a significant consideration in the design of desired optical devices. For example, the composition and properties, such as density, of a material can be adjusted to obtain materials with a desired index-of-refraction. Similarly, the thermal expansion and flow temperatures of a material can be consistent with a reasonable processing approach for forming the materials into a monolithic, integrated structure. The consolidated optical materials can have good optical properties such that light transmission through the materials does not result in undesirable amount of loss. In addition, the materials generally should have properties that allow processing under reasonable conditions to form the integrated devices of the integrated optical circuit or electro-optical circuit. The formation of integrated optical devices using light reactive deposition is described further in copending and commonly assigned U.S. Patent Application serial number 10/027,906 to Bi et al., ^{not pending} entitled "Three Dimensional Engineering of Optical Structures," incorporated herein by reference.

The materials described herein, e.g., doped glasses, can be useful in the production of optical devices. Using the techniques described herein, the materials can be formulated into planar optical devices. The composition of the material, including any dopant/additive, can change the optical properties of the materials to be suitable for particular optical applications. Materials of particular interest herein include, for example, amplifying materials, paramagnetic materials and photosensitive materials.

Rare earth doped glasses are particularly suitable for use in the formation of optical amplifiers. The amplifier material can be excited by a pump light signal coupled to the optical material. The pump light excites the rare earth doped materials. A optical input passing through the

Ce, Tb, Dy, Pr and Tm. Suitable non-rare earth metal dopants/additives include, for example, Al, Ga, Mg, Sr, Zn, Bi, Sb, Zr, Pb, Li, Na, K, Ba, W, Si, Ge, P, B, Te and Ca.

Material processing remains a significant consideration in the design of desired optical devices. For example, the composition and properties, such as density, of a material are adjusted to obtain materials with a desired index-of-refraction. Similarly, the thermal expansion and flow temperatures of a material have to be consistent with a reasonable processing approach for forming the materials into a monolithic, integrated structure without excessive stress that can introduce undesirable optical properties such as unintentional birefringence. The consolidated optical materials can provide high transparency and homogeneity at the operating wavelength such that light transmission through the materials does not result in undesirable amount of loss. In addition, the materials have to be processable under reasonable conditions to form the integrated devices of the integrated optical circuit or electro-optical circuit. Similar material constraints can be problematic for the formation of state-of-the-art integrated electronic devices.

Various materials have been formed as submicron/nanoscale particles using laser pyrolysis. Some of these materials are described in the following description. Using light reactive deposition, these materials can be formed directly as coatings in the form of powder arrays. Based on the description and examples herein, a range of additional materials can be produced by laser pyrolysis and light reactive deposition. Specifically, suitable approaches for the formation of some improved materials are outlined below.

For example, the production of silicon oxide nanoparticles is described in copending and commonly assigned U.S. Patent Application Serial Number 09/085,514 to Kumar et al., ^{now US Patent No. 6,726,980} entitled "Silicon Oxide Particles," incorporated herein by reference. This patent application describes the production of amorphous SiO₂. The production of titanium oxide nanoparticles and crystalline silicon dioxide nanoparticles is described in copending and commonly assigned, U.S. Patent Application Serial Number 09/123,255 to Bi et al., ^{now US Patent No. 6,387,531} entitled "Metal (Silicon) Oxide/Carbon Composites," incorporated herein by reference. In particular, this application describes the production of anatase and rutile TiO₂.

In addition, nanoscale manganese oxide particles have been formed. The production of these particles is described in copending and commonly assigned U.S. Patent Application Serial No. 09/188,770 to Kumar et al., ^{now US Patent No. 6,506,493} entitled "Metal Oxide Particles," incorporated

herein by reference. This application describes the production of MnO , Mn_2O_3 , Mn_3O_4 and Mn_5O_8 .

Also, the production of vanadium oxide nanoparticles is described in U.S. Patent 6,106,798 to Bi et al., entitled "Vanadium Oxide Nanoparticles," incorporated herein by reference.

5 Similarly, silver vanadium oxide nanoparticles have been produced, as described in U.S. Patent 6,225,007 to Horne et al., and copending and commonly assigned U.S. Patent Application Serial No. 09/311,506 to Reitz et al., ^{now USP No 6,391,494} both entitled "Metal Vanadium Oxide Particles," both of which are incorporated herein by reference.

10 Furthermore, lithium manganese oxide nanoparticles have been produced by laser pyrolysis along with or without subsequent heat processing, as described in copending and commonly assigned U.S. Patent Applications Serial No. 09/188,768 to Kumar et al., ^{now USP No 6,607,706} entitled "Composite Metal Oxide Particles," and 09/334,203 to Kumar et al., ^{now USP No 6,482,374} entitled "Reaction Methods for Producing Ternary Particles," and U.S. Patent 6,136,287 to Horne et al., entitled "Lithium Manganese Oxides and Batteries," all three of which are incorporated herein by reference.

15 The production of aluminum oxide nanoparticles is described in copending and commonly assigned, U.S. Patent Application Serial Number 09/136,483 to Kumar et al., ^{now pending} entitled "Aluminum Oxide Particles," incorporated herein by reference. In particular, this application disclosed the production of $\gamma\text{-Al}_2\text{O}_3$. The formation of $\delta\text{-Al}_2\text{O}_3$ and $\theta\text{-Al}_2\text{O}_3$ by laser pyrolysis/light reactive deposition along with doped-crystalline and amorphous alumina is
20 described in copending and commonly assigned U.S. Patent Application serial number 09/969,025 to Chiruvolu et al., ^{now pending} entitled "Aluminum Oxide Powders," incorporated herein by reference.

Amorphous aluminum oxide materials can be combined with other glass formers, such as SiO_2 and/or P_2O_3 . For example, suitable metal oxide dopants/additives for aluminum oxide
25 for optical glass formation comprise cesium oxide (Cs_2O), rubidium oxide (Rb_2O), thallium oxide (Tl_2O), lithium oxide (Li_2O), sodium oxide (Na_2O), potassium oxide (K_2O), beryllium oxide (BeO), magnesium oxide (MgO), calcium oxide (CaO), strontium oxide (SrO), barium oxide (BaO), and the like and suitable combinations of any two or more thereof. Glass dopants/additives can affect, for example, the index-of-refraction, consolidation temperature and/or the porosity of the glass.
30 Suitable metal oxide dopants/additives for infrared emitters comprise, for example, cobalt oxide

(Co₃O₄), Er₂O₃, CrO₂, Tm₂O₃, Nd₂O₃, Yb₂O₃, Pr₂O₃, Dy₂O₃, Ho₂O₃, and the like, and suitable combinations of any two or more thereof.

In addition, tin oxide nanoparticles have been produced by laser pyrolysis, as described in U.S. Patent 6,200,674 to Kumar et al., entitled "Tin Oxide Particles," incorporated
5 herein by reference. The production of zinc oxide nanoparticles is described in copending and commonly assigned U.S. Patent Application Serial Number 09/266,202 to Reitz, ^{now pending} entitled "Zinc Oxide Particles," incorporated herein by reference. In particular, the production of ZnO nanoparticles is described.

Submicron and nanoscale particles and corresponding coatings of rare earth metal
10 oxide particles, rare earth doped metal/metalloid oxide particles, rare earth metal/metalloid sulfides and rare earth doped metal/metalloid sulfides are described in copending and commonly assigned U.S. Patent Application serial number 09/843,195 to Kumar et al., ^{now USP No 6,692,660} entitled "High Luminescence Phosphor Particles," incorporated herein by reference. Suitable host materials for the formation of phosphors comprise ZnO, ZnS, Zn₂SiO₄, SrS, YBO₃, Y₂O₃, Al₂O₃, Y₃Al₅O₁₂ and BaMgAl₁₄O₂₃,
15 and suitable combinations of any two or more thereof. Exemplary non-rare earth metals for activating phosphor particles as dopants/additives include, for example, manganese, silver and lead. Exemplary rare earth metals for forming metal oxide phosphors include, for example, europium, cerium, terbium and erbium. Generally, heavy metal ions or rare earth ions are used as activators in phosphors. For phosphor applications, the particles are generally crystalline.

The production of iron, iron oxide and iron carbide is described in a publication
20 by Bi et al., entitled "Nanocrystalline α -Fe, Fe₃C, and Fe₇C₃ produced by CO₂ laser pyrolysis," J. Mater. Res. Vol. 8, No. 7 1666-1674 (July 1993), incorporated herein by reference. The production of nanoparticles of silver metal is described in copending and commonly assigned U.S. Patent Application Serial Number 09/311,506 to Reitz et al., ^{now USP No 6,381,494} entitled "Metal Vanadium
25 Oxide Particles," incorporated herein by reference. Nanoscale carbon particles produced by laser pyrolysis is described in a reference by Bi et al., entitled "Nanoscale carbon blacks produced by CO₂ laser pyrolysis," J. Mater. Res. Vol. 10, No. 11, 2875-2884 (Nov. 1995), incorporated herein by reference.

The production of iron sulfide (Fe_{1-x}S) nanoparticles by laser pyrolysis is
30 described in Bi et al., Material Research Society Symposium Proceedings, vol. 286, p. 161-166

(1993), incorporated herein by reference. Precursors for laser pyrolysis production of iron sulfide were iron pentacarbonyl ($\text{Fe}(\text{CO})_5$) and hydrogen sulfide (H_2S). Other suitable gaseous sulfur precursors for vapor delivery comprise, for example, pyrosulfuryl chloride ($\text{S}_2\text{O}_5\text{Cl}_2$), sulfur chloride (S_2Cl_2), sulfuryl chloride (SO_2Cl_2), thionyl chloride (SOCl_2), and the like, and suitable combinations of any two or more thereof. Suitable sulfur precursors for aerosol delivery comprise, for example, ammonium sulfate ($(\text{NH}_4)_2\text{S}$), sulfuric acid (H_2SO_4), and the like, and any suitable combinations thereof, which are soluble in water. Other metal/metalloid sulfide materials can be similarly produced.

Cerium oxide can be produced using the laser pyrolysis apparatuses described above. Suitable precursors for aerosol delivery comprise, for example, cerous nitrate ($\text{Ce}(\text{NO}_3)_3$), cerous chloride (CeCl_3), cerous oxalate ($\text{Ce}_2(\text{C}_2\text{O}_4)_3$), and the like, and suitable combinations of any two or more thereof. Similarly, zirconium oxide can be produced using the laser pyrolysis apparatuses described above. Suitable zirconium precursors for aerosol delivery comprise, for example, zirconyl chloride (ZrOCl_2), zirconyl nitrate ($\text{ZrO}(\text{NO}_3)_2$), and the like, and suitable combinations of any two or more thereof.

The deposition of coatings of dielectric materials for chip capacitors is described in copending and commonly assigned U.S. Provisional Patent Application serial number 60/312,234 to Bryan, entitled "Reactive Deposition For The Formation Of Chip Capacitors," incorporated herein by reference. Suitable dielectric materials include a majority of barium titanate (BaTiO_3), optionally mixed with other metal oxides. Other dielectric oxides suitable for incorporation into ceramic chip capacitors with appropriate dopants/additives comprise, for example, SrTiO_3 , CaTiO_3 , SrZrO_3 , CaZrO_3 , $\text{Nd}_2\text{O}_3\text{-}2\text{TiO}_3$, $\text{La}_2\text{O}_3\text{-}2\text{TiO}_2$, and the like, and any two or more thereof.

The production of ternary nanoparticles of aluminum silicate and aluminum titanate can be performed by laser pyrolysis following procedures similar to the production of silver vanadium oxide nanoparticles described in copending and commonly assigned U.S. Patent Application Serial Number 09/311,506 to Reitz et al., ^{now U.S. Patent No. 6,391,494} entitled "Metal Vanadium Oxide Particles," incorporated herein by reference. Suitable precursors for the production of aluminum silicate comprise, for vapor delivery, a mixture of aluminum chloride (AlCl_3), silicon tetrachloride (SiCl_4), and the like, and suitable combinations of any two or more thereof, and, for aerosol delivery, a mixture of tetra(N-butoxy) silane and aluminum isopropoxide

(Al(OCH(CH₃)₂)₃), a mixture of tetraethoxysilane and aluminum nitrate, or tetraethoxysilane and aluminum chloride, and the like, and suitable combinations of any two or more thereof. Similarly, suitable precursors for the production of aluminum titanate comprise, for aerosol delivery, a mixture of aluminum nitrate (Al(NO₃)₃) and titanium dioxide (TiO₂) powder dissolved in sulfuric acid, a mixture of aluminum isopropoxide and titanium isopropoxide (Ti(OCH(CH₃)₂)₄), and the like, and suitable combinations of any two or more thereof.

The formation of submicron and nanoscale particles along with coatings of metal/metalloid compositions with complex anions is described in copending and commonly assigned U.S. Patent Application serial number 09/845,985 to Chaloner-Gill et al., ^{now pending} entitled

10 "Phosphate Powder Compositions And Methods For Forming Particles With Complex Anions," incorporated herein by reference. Suitable polyatomic anions comprise, for example, phosphate (PO₄⁻³), sulfate (SO₄⁻²), silicate (SiO₄⁻⁴), and the like, and suitable combinations of any two or more thereof. Suitable phosphorous precursors for forming the phosphate anion, sulfur precursors for forming the sulfate anion and silicon precursors for forming the silicate anion are discussed above. Suitable cations comprise, for example, metal and metalloid cations. Phosphate glasses can be used in a variety of contexts. Phosphate compositions for glasses comprise, for example, aluminum phosphate (AlPO₄), calcium phosphate (Ca₃(PO₄)₂), and the like, and suitable combinations of any two or more thereof. Suitable gaseous phosphate precursor compositions for vapor delivery comprise, for example, phosphine (PH₃), phosphorus trichloride (PCl₃), phosphorous pentachloride (PCl₅), phosphorus oxychloride (POCl₃), P(OCH₃)₃, and the like, and suitable combinations of any two or more thereof. Suitable phosphorous precursors for aerosol delivery comprise, for example, (C₂H₅O)₃P, (C₂H₅O)₃PO, ammonium phosphate ((NH₄)₃PO₄), ammonium phosphate - dibasic ((NH₄)₂HPO₄), ammonium phosphate - monobasic ((NH₄)H₂PO₄), phosphoric acid (H₃PO₄), and the like, and suitable combinations of any two or more thereof, which are all moderately soluble in water.

The synthesis by laser pyrolysis of silicon carbide and silicon nitride is described in copending and commonly assigned U.S. Patent Application serial No. 09/433,202 to Reitz et al., ^{now pending} entitled "Particle Dispersions," incorporated herein by reference. Other metal/metalloid carbides and meta/metalloid nitrides can be similarly produced.

condensed aerosol from outer nozzle 212. Inner nozzle 214 is secured to outer nozzle 212 at fitting 224.

The top of inner nozzle 214 can be a twin orifice internal mix atomizer 226. Liquid is fed to the atomizer through tube 228, and gases for introduction into the reaction chamber are fed to the atomizer through tube 230. Interaction of the gas with the liquid assists with droplet formation.

A plurality of aerosol generators can be used to produce aerosol within the reaction chamber or within one or more inlets leading to the reaction chamber. The aerosol generators can be used to generate the same or different aerosol composition from each other. For embodiments in which the aerosol generators product aerosols of different compositions, the aerosols can be used to introduce reactants/precursors that are not easily or conveniently dissolved/dispersed into the same solvent/dispersant. Thus, if a plurality of aerosol generators is used to form an aerosol directly within the reaction chamber, the aerosol generators can be oriented to mix the reactants or to deliver separate streams, possibly overlapping, along the reaction zone. If two or more aerosols are generated within a single inlet nozzle the aerosols can be mixed and flowed within a common gas flow. An inlet nozzle with two aerosol generators is shown in Fig. 3B. Inlet nozzle 240 includes aerosol generators 242, 244. Which generate aerosols directed to outlet 246.

Alternatively, aerosol generators can generate aerosols within separate inlets such that the aerosols are combined within the reaction chamber. The use of a plurality of aerosol generators within a single inlet nozzle or a plurality of inlet nozzles can be useful for embodiments in which it is difficult to introduce desired compositions within a single solution/dispersion. Multiple aerosol generators producing aerosols within different inlets is described further in copending and commonly assigned U.S. Patent application serial number 09/362,631 to Mosso et al., *now pending*, entitled "Particle Production Apparatus," incorporated herein by reference.

In any of these aerosol embodiments, one or more vapor/gas reactants/precursors can also be introduced. For example, the vapor/gas precursors can be introduced within the aerosol generator itself to help form the aerosol. In alternative embodiments, the vapor can be delivered through a separate inlet into the delivery channel into which the aerosol is generated such that the vapor and aerosol mix and are delivered into the reaction chamber through the same reactant inlet. In further embodiments, the vapor precursors are delivered into the reaction chamber through

26
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and commonly assigned U.S. Patent Application Serial No. 09/362,631 to Mosso et al., entitled "Particle Production Apparatus," incorporated herein by reference.

In one embodiment of a commercial capacity laser pyrolysis apparatus, the reaction chamber and reactant inlet are elongated significantly along the light beam to provide for an increase in the throughput of reactants and products. The embodiments described above for the delivery of aerosol reactants can be adapted for the elongated reaction chamber design. Additional embodiments for the introduction of an aerosol with one or more aerosol generators into an elongated reaction chamber are described in U.S. Patent 6,193,936 to Gardner et al., entitled "Reactant Delivery Apparatuses," incorporated herein by reference. A combination of vapor and aerosol precursors can be delivered into this reaction chamber by generalizing the approaches discussed above with respect to Figs. 3-5. These improved reactors and corresponding nozzles can be adapted for light reactive deposition with vapor precursors, aerosol precursors and combinations thereof.

In general, the laser pyrolysis apparatus with the elongated reaction chamber and reactant inlet is designed to reduce contamination of the chamber walls, to increase the production capacity and to make efficient use of resources. To accomplish these objectives, the elongated reaction chamber provides for an increased throughput of reactants and products without a corresponding increase in the dead volume of the chamber. The dead volume of the chamber can become contaminated with unreacted compositions and/or reaction products. Furthermore, an appropriate flow of shielding gas confines the reactants and products within a flow stream through the reaction chamber. The high throughput of reactants makes efficient use of the laser energy.

The design of the improved reaction chamber 472 is shown schematically in Fig. 9. A reactant inlet 474 leads to main chamber 476. Reactant inlet 474 conforms generally to the shape of main chamber 476. Main chamber 476 includes an outlet 478 along the reactant/product stream for removal of particulate products, any unreacted gases and inert gases. The configuration can be reversed with the reactants supplied from the top and product collected from the bottom, if desired. Shielding gas inlets 480 are located on both sides of reactant inlet 474. Shielding gas inlets are used to form a blanket of inert gases on the sides of the reactant stream to inhibit contact between the chamber walls and the reactants or products. The dimensions of elongated main chamber 476 and reactant inlet 474 can be designed for high efficiency particle production. Reasonable lengths for

The temperature of the substrate during the deposition process can be adjusted to achieve particular objectives. For example, the substrate can be cooled during the deposition process since a relatively cool substrate can attract the particles to its surface. However, in some embodiments, the substrate is heated, for example to about 500°C, during the deposition process.

5 Particles stick better to a heated substrate. In addition, the particles tend to compact and fuse on a heated substrate such that a subsequent consolidation of the coating into a fused glass or other material is facilitated if the coating were formed initially on a heated substrate.

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10 The formation of coatings by light reactive deposition, silicon glass deposition and optical devices in general are described further in copending and commonly assigned U.S. Patent Application 09/715,935 to Bi et al., ^{now pending} entitled "Coating Formation By Reactive Deposition," incorporated herein by reference, and in copending and commonly assigned PCT application designating the U.S. serial number PCT/US01/32413 to Bi et al. filed on October 16, 2001, entitled "Coating Formation By Reactive Deposition," incorporated herein by reference.

15 The well-defined reactant stream as a sheet of flow leading into the reaction zone tends to spread after the reaction zone due to heat from the reaction. If the substrate is swept through the reaction zone near the reaction zone, the spreading of the flow may not be significant. In some embodiments, it may be desirable to sweep the substrate through the flow farther away from the reaction zone such that the flow has spread significantly and the entire substrate or desired portion thereof can be coated simultaneously without moving the substrate.
20 The appropriate distance to obtain a uniform coating of particles depends on the substrate size and the reaction conditions. A typical distance of about 15 centimeters would be suitable for a wafer with a 4-inch diameter.

25 In embodiments in which the entire substrate surface is simultaneously coated, when the composition of the product particle flow is changed in time during the deposition process, the composition of the particles changes through the thickness of the coating. If the composition is changed continuously, a continuous composition gradient through the layer results. For optical materials, generally a continuous composition gradient layer comprising a continuous composition change from a first composition to a second composition has a thickness of no more than about 300 microns, in other embodiments no more than about 150 microns, in
30 further embodiments, in the range(s) from about 500 nm to about 100 microns and in still other

be removed intact from the surface that is coated. Therefore, the discrete mask approach herein is different from previous masking approaches adapted from photolithography for vapor deposition approaches.

In these embodiments, the formation of the particle coating correspondingly involves directing a product particle stream at the substrate shielded with the discrete mask. The discrete mask has a surface, generally a planar surface, with openings at selected locations. The discrete mask blocks the surface except at the openings such that particles can deposit on the surface through the openings. Thus, the mask provides for patterning compositions on the surface by the selected placement of the openings. In some embodiments, suitable discrete masks comprise a mask with a slit that is narrower than the product particle flow such that the deposition process can be very precisely controlled. Movement of the slit can form a desired, precisely controlled pattern with one or more compositions. After use of a discrete mask, it can be removed and reused.

In some embodiments, a plurality of masks can be used to deposit particles along a single layer. For example, following deposition of a pattern through a first mask, a second complementary mask can be used to deposit material over at least a portion of the surface left uncovered during deposition with the first mask. Further complementary masks can be used to form complex patterns while completing a single layer or portion thereof with a coating having varying chemical composition over the layer.

Thus, using light reactive deposition, a range of effective approaches are available to vary the chemical composition of optical materials within layers and in different layers to form three-dimensional optical structures with selected compositions are selected locations within the material. The patterning of compositions of optical materials during the deposition process is described further in copending and commonly assigned U.S. Patent application 10/027,906 to Bi et al., ^{now pending} entitled "Three Dimensional Engineering of Optical Structures," incorporated herein by reference.

As described in detail above, laser pyrolysis apparatuses and corresponding light reactive deposition apparatuses have been designed for the production of commercial quantities of nanoscale powders and powder arrays. Alternatively or in addition, the invention provides that the rate of production and/or deposition of the particles can be varied substantially,

light. This provides for the unidirectional propagation of light within a core waveguide. The magnitude of the Verdet constant and therefore the Faraday effect can be increased by enhancing the paramagnetism. An enhanced Faraday effect can be used to construct smaller optical devices, such as optical rotators and Faraday isolators

5 Layered structures for optical fiber preforms and planar optical structures, including integrated planar optical structures, are described further in copending and commonly assigned PCT application designating the U.S. PCT/01/45762 to Bryan et al., entitled "Multilayered Optical Structures," incorporated herein by reference and copending and commonly assigned U.S. Patent Application serial number 10/027,906 to Bi et al. ^{now pending} entitled "Three Dimensional Engineering Of
10 Optical Structures," incorporated herein by reference.

EXAMPLES

Example 1 - Erbium Doped Silica Glass

This example describes the coating of a silicon substrate with a silica glass including alumina and sodium oxide glass formers and an erbium dopant using light reactive deposition and consolidation.

Particle coating was performed using light reactive deposition in which wafer coating was performed within the reaction chamber by sweeping the substrate through a product particle stream. The wafer was a silicon wafer with a thermal oxide under-cladding. The apparatus used to coat a substrate/wafer in the reaction stream is comparable to the apparatus shown in Figs. 20-22 with an aerosol precursor delivery system similar the system shown schematically in Fig. 5. The coating was performed with a static coating configuration. An oxygen/ethylene flame was started first. Then, the aerosol flow was started. When a stable process flame was observed, the wafer was translated into the coating position about 17 inches above the laser beam. At this distance, the product particle flow has spread such that the entire surface is simultaneously coated approximately uniformly. The wafer was left in the flow for about 20 minutes.

A solution was formed combining 66 grams (g) tetraethoxysilane ($\text{Si}(\text{OC}_2\text{H}_5)_4$ or TEOS, 99.9% pure), 25.6 g aluminum nitrate ($\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, >98% pure), 9.5 g sodium nitrate (NaNO_3 , 99% pure), and 1.9 g erbium nitrate ($\text{Er}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$, 99.99% pure) in a isopropyl